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Fission Product Radioactivity in the Air Along the 80th Meridian (West) During 1961

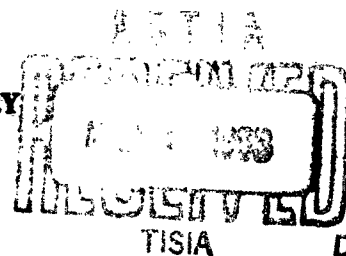
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The concentrations of fission product radioactivity in the atmosphere continued the trends exhibited since the beginning of the moratorium on nuclear testing; namely, a clearly defined seasonal variation in activity at sites in the northern hemisphere, together with a less obvious one in the southern hemisphere, and a tendency toward equilibration of the activity concentrations in the two hemispheres, through both a slowly decreasing concentration of fission products in the atmosphere north of the equator and an increase south of the equator. Prior to reinitiation of the Soviet nuclear testing program in September 1961, the natural radioisotope Pb^{210} had become a significant contributor to the total long-lived β activity in the atmosphere. After September 1961, the fission product radioactivity in the northern hemisphere increased a hundredfold or more; a small but measurable transfer of activity to the southern hemisphere occurred during the last months of 1961.

INTRODUCTION

The cooperative 80th meridian air sampling program, which was initiated in 1956 and carried out during 1957-1958 and 1958-1959 as an integral part, respectively, of the International Geophysical Year (IGY) and International Geophysical Cooperation (IGC-59) programs on atmospheric nuclear radiation, has continued to actively monitor the atmosphere at ground level for the presence of fission product radioactivity. This earlier work is reported elsewhere (1-7).

The purpose of this network of stations is to provide a means of studying the effect of latitude on the distribution of fission products in the atmosphere, to obtain information on the mechanism and rate of mixing of fission debris in a north-south direction and particularly across the equator, and to serve as a reference line to which the various independent measurements of fission product concentrations can be related.

During the year 1961 air sampling was performed at 13 sites essentially along the 80th meridian (west) as the result of the interest and cooperation of various agencies in the countries located along this meridian (Table 1). An additional station at Mauna Loa, Hawaii, though far from the 80th meridian, has been operated in conjunction with this network because it is located at a high elevation in the northern hemisphere equivalent to that of the high altitude station at Chacaltaya in the southern hemisphere.

Sampling was conducted on a weekly basis during 1961, rather than on the thrice-weekly schedule employed in 1960 or the daily schedule employed in earlier years, because of the generally decreased fission product activity present in the

air and the desirability of having statistically significant counting rates in the individual samples. This weekly schedule was maintained after the resumption of testing by the Soviet Union; since the networks of the U.S. Public Health Service and of the Canadian Department of Health and Welfare provided information on arrival times of debris at various localities, it was decided that insufficient additional information would have been obtained to have warranted resuming a daily sampling procedure at each 80th meridian site.

EXPERIMENTAL PROCEDURE

The procedure employed in the 80th meridian air sampling program involved the collection of airborne particulate matter by drawing ground-level air through highly efficient air filters (cellulose-asbestos Type 6) 8 inches in diameter by the use of positive-displacement blowers at the rate of approximately 1200 cubic meters per day. Identical sampling equipment was employed at each site; corrections have been applied to the airflow rate to take into account the effect of the station altitudes on the efficiency of the blower, and also to take into account the blower rotation rate, which is a function of the frequency of the alternating current supply. Filters were normally changed each Monday after 7 days exposure; they were returned to NRL by air for assay for gross β activity and subsequently for radiochemical analysis.

The samples were ashed by heating to 650°C in a muffle furnace, the ash compressed into discs 1½ inches in diameter and counted under an end-window Geiger counter (both flow type and organic-quenched counter tubes employed) in an automatic sample changer two weeks after the end of the collections. Low activity samples were counted on a lead and anticoincidence shielded

TABLE 1
Collecting Sites Associated with the NRL 80th Meridian
Air Sampling Program During 1961

Station	Latitude	Longitude	Elevation	Operator
Thule, Greenland	76°35'N	68°35'W	259	Geophysics Research Directorate, USAF, Cambridge Research Laboratories
Moosonee, Ontario, Canada	51°16'N	89°39'W	10	Meteorological Branch, Department of Transport (Canada)
Washington, D.C.	38°59'N	77°29'W	82	U. S. Weather Bureau
Miami, Florida	25°49'N	80°17'W	4	U. S. Weather Bureau
Mauna Loa, Hawaii	19°28'N	155°36'W	3394	U. S. Weather Bureau
San Juan, Puerto Rico	18°26'N	66°00'W	10	U. S. Weather Bureau
Miraflores, Panama Canal Zone	9°00'N	79°35'W	10	Canal Zone Corrosion Laboratory (U. S. Naval Research Laboratory)
Guayaquil, Ecuador	2°10'S	79°52'W	7	Meteorological Office, Direccion General de Aviacion Civil
Lima, Peru*	12°01'S	77°07'W	30	Corporacion Peruana de Aeropuertos y Aviacion Comercial (CORPAC)
Chacaltaya, Bolivia	17°10'S	68°15'W	5220	Universidad Mayor de San Andres, Laboratorio de Fisica Cosmica de Chacaltaya
Antofagasta, Chile	23°37'S	70°16'W	519	NASA Satellite Tracking Station
Santiago, Chile	33°27'S	70°42'W	520	Oficina Meteorologica de Chile
Puerto Montt, Chile	41°27'S	72°57'W	5	Oficina Meteorologica de Chile
Punta Arenas, Chile	53°08'S	70°53'W	3	Oficina Meteorologica de Chile

*Collecting equipment moved from 12°06'S, 77°01'W on 3 July 1961.

counter having a background of three counts/minute and an efficiency toward RaE of 11.4%; the more active samples were counted in a lead-shielded counter having a background of ten counts/minute and an efficiency of 3.6%. The time to accumulate a preset count of 2000 was measured; generally, however, when the activity was sufficiently high, five or more cycles through the series of samples was permitted with a total accumulated count of 10,000 or more per sample. Each counting cycle also included the measurement of the counter background and a radioactive standard.

Counting efficiency was determined relative to a known activity of RaE(Bi²¹⁰) in an equilibrium mixture of RaDEF mounted with one gram of

filter ash. A series of such standards was employed to calibrate for the varying weights of filter ash (generally in the range 0.7 to 1.2 grams per sample) resulting from variations in filter composition and quantity of accumulated dust.

Radiochemical analyses were performed bi-monthly on samples from each site (except San Juan, Puerto Rico) for the following radioisotopes: Sr⁸⁹, Sr⁹⁰, Y⁹¹, Cs¹³⁷, Ce¹⁴¹, Ce¹⁴⁴, Pm¹⁴⁷, and Pb²¹⁰. Counting techniques were standardized by counting samples of radioisotopes having known disintegration rates under the identical conditions (sample size and weight, counter geometry) used to evaluate the isotopes separated from the filter ash. The isotopes employed as standards were C¹⁴, Co⁶⁰, Cs¹³⁷, Sr⁹⁰, Tl²⁰⁴, Bi²¹⁰ (RaE), Y⁹⁰, and Pa²³⁴ (UX₂

in equilibrium with U^{238} in natural U_3O_8). For isotopes having β_{max} energies differing from the above, correction factors were read from calibration curves obtained from plots of the β_{max} energy versus the counting efficiency of the various standards.

In the case of elements having two or more isotopes contributing to the total activity of the separated carrier element, differentiation was made by one or more of the following methods: (a) Y^{90} was milked from the Sr^{90} - Sr^{90} (Y^{90}) mixture and both the Y^{90} and Sr^{90} - Sr^{90} fractions counted, after which a correction was applied for the Sr^{90} contribution to the Sr^{90} - Sr^{90} fraction to obtain the Sr^{90} counting rate; (b) Ce^{141} - Ce^{144} (Pr^{144}) samples were counted without an absorber and again through a 0.025-inch aluminum absorber, which was sufficient to absorb completely the β particles from Ce^{141} and Ce^{144} , after which the contribution of each component could be calculated; the Pm^{147} and Y^{91} contributions to the activity of the separated yttrium carrier were determined in a similar manner; and (c) samples were recounted after additional decay had taken place and the contribution of the isotopes determined from the decrease in total count occurring during this period; this method was applicable to Sr^{90} - Sr^{90} (Y^{90}), Ce^{141} - Ce^{144} (Pr^{144}), and Y^{91} - Pm^{147} samples, in which isotopes of widely differing half-lives were involved.

GROSS FISSION PRODUCT CONCENTRATIONS IN THE AIR

Monthly averages of the gross fission product concentrations found in the air at ground level at the various sampling sites are reported in Table 2, and are shown graphically in Fig. 1 as latitudinal profiles of radioactivity. These results may be compared with those reported by the Radiation Surveillance Network of the U.S. Public Health Service and by the Air Sampling Program of the Radiation Protection Division, Canadian Department of National Health and Welfare, by multiplying the 80th meridian air concentrations by the experimentally determined factors of 0.58 and 0.87, respectively (8).

In the northern hemisphere the gross β activity levels had decreased during 1961 to nearly one-half the values for similar periods in 1960; in the southern hemisphere the decrease was not quite as large. The decrease was due both to deposition of radioactivity and to radioactive decay, though this latter effect was becoming of less importance with the passage of time since the conclusion of nuclear testing in 1958. Radiochemical analyses showed that in many areas a large contributor to the gross β activity was the long-lived natural radioisotope Pb^{210} (RaD) and its daughter Bi^{210} (RaE).

TABLE 2
Gross Fission Products in the Ground-Level Air During 1961

Activity (dis/min/m ³ of air)												
Site	Jan.	Feb.	Mar.	Apr.	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.
Thule, Greenland	0.11	0.13	0.14	0.26	0.14	0.17	0.08	0.05	2.32	3.82	11.8	13.0
Moosonee, Ontario	0.11	—	0.23	—	0.24	0.16	0.10	0.06	10.3	5.92	11.4	5.21
Washington, D.C.	0.13	0.15	0.19	0.23	0.31	0.33	0.18	0.13	7.86	9.40	11.4	9.31
Miami, Florida	0.18	0.20	0.28	0.41	0.29	0.14	0.10	0.05	3.37	6.61	13.5	16.4
Mauna Loa, Hawaii	0.08	0.08	0.16	0.20	0.20	0.19	0.13	0.06	0.63	1.27	7.39	8.35
San Juan, P.R.	0.23	—	—	0.12	0.21	—	—	—	—	0.79	3.53	5.40
Miraflores, P.C.Z.	0.60	0.11	0.15	0.12	0.16	0.05	0.03	0.03	0.03	0.11	2.31	3.17
Guayaquil, Ecuador	0.05	0.04	0.04	0.05	0.04	0.04	0.01	0.02	0.02	0.03	0.08	0.18
Lima, Peru	0.04	0.04	0.03	0.03	0.05	0.07	0.06	0.05	0.07	0.07	0.06	0.06
Chacaltaya, Bolivia	0.03	0.03	0.04	0.03	0.03	0.04	0.06	0.07	0.04	0.08	0.09	0.08
Antofagasta, Chile	0.09	0.14	0.08	0.08	0.08	0.07	0.07	0.06	0.08	0.08	0.10	0.20
Santiago, Chile	0.12	—	—	—	0.08	—	—	0.06	0.07	0.06	0.08	0.11
Puerto Montt, Chile	—	—	0.05	—	—	0.03	0.04	0.04	0.03	0.04	0.03	0.06
Punta Arenas, Chile	0.03	0.04	0.03	0.03	0.02	0.03	0.03	0.03	0.02	0.02	—	0.03

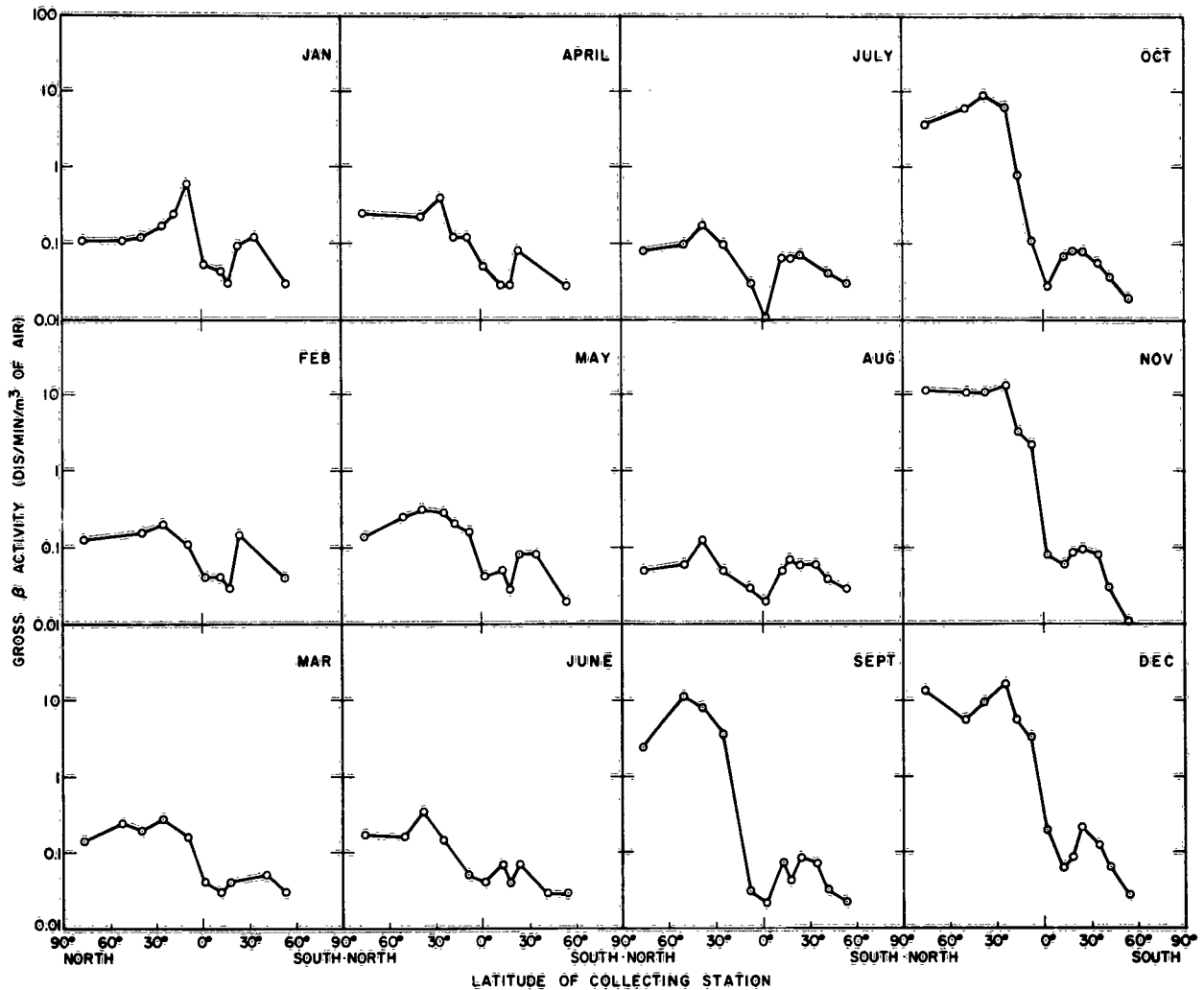


Fig. 1 - Profiles of the gross fission product concentrations in the ground-level air along the 80th meridian (west) during 1961

The only effect of the French nuclear testing program on the gross β activity of the air was an observed increase in activity over a short period in mid-January at Miraflores, Panama Canal Zone, and a lesser increase at San Juan, Puerto Rico, following the December 27, 1960, test in the Sahara. This is evident in the radioactivity profile for January (Fig. 1); the more sensitive and specific radiochemical analyses indicated that a trace of this fresh activity did also appear at Guayaquil, Ecuador. There was no evidence in either the gross activity measurements or in the radiochemical analyses that debris from the French nuclear test of April 25, 1961, appeared at any 80th meridian site.

The extensive contamination of the atmosphere of the northern hemisphere as the result of the resumption of nuclear testing by the Soviet Union is

evident in September 1961 and in the following months. This fresh activity only slowly penetrated southward and it was not until November that any quantity of it appeared as far south as Panama (9°N). Though the gross β measurements did not clearly indicate the presence of any of this activity in the southern hemisphere until early 1962, radiochemical analysis indicated some transfer of activity across the equator during the period November-December 1961 and possibly earlier (Table 3). One difficulty in associating any increase in gross β activity in the southern hemisphere with an intrusion of fresh debris is that during this period the normal seasonal rise in deposition of older debris held in the stratosphere is taking place.

In spite of the introduction of fresh debris by the French and Soviet tests, the normal patterns of increases in activity (maxima) during the spring

TABLE 3

Month	Days** Sampled	Activity (dis/min per 100 std. cubic meters of air)†									Activity Ratios							
		Gross β	Ce ¹⁴¹	Sr ⁹⁰	Y ⁹¹	Ce ¹⁴⁴	Pm ¹⁴⁷	Sr ⁹⁰	Ce ¹³⁷	Pb ²¹⁰	Ce ¹⁴¹ / Ce ¹⁴⁴	Ce ¹⁴¹ / Pm ¹⁴⁷	Ce ¹⁴⁴ / Sr ⁹⁰	Ce ¹³⁷ / Sr ⁹⁰	Pb ²¹⁰ / Sr ⁹⁰	Pb ²¹⁰ / Gross β	Sr ⁹⁰ / Sr ⁹⁰	
Thule, Greenland - Lat. 76°35'N Long. 68°35'W Elev. 259 m																		
Jan-Feb	59	12	*	*	*	1.49	1.54	0.489	0.867	2.27	0.0	0.97	3.05	1.77	4.6	0.19	0.0	
Mar-Apr	59	20	*	*	*	-	-	0.974	1.65	2.75	-	-	-	1.69	2.8	0.14	0.0	
May-June	63	16	*	*	*	2.13	2.20	0.722	1.42	1.51	0.0	0.97	2.95	1.97	2.1	0.09	0.0	
July-Aug	59	7	*	*	*	0.823	1.07	0.350	0.615	0.731	0.0	0.77	2.35	1.76	2.1	0.10	0.0	
Sept-Oct	63	307	67.3	25.4	35.5	10.6	2.62	0.312	0.453	0.797	6.35	4.05	34.0	1.45	2.6	0.0026	81.4	
Nov-Dec	58	1240	249.	125.	197.	76.4	-	2.05	2.65	2.05	3.26	-	37.3	1.29	1.0	0.0017	61.0	
Moosonee, Ontario, Canada - Lat. 51°16'N Long. 80°39'W Elev. 10 m																		
Jan	25	11	*	*	*	0.930	0.784	0.294	0.465	4.30	0.0	1.19	3.16	1.58	14.6	0.39	0.0	
Mar	32	23	*	*	*	2.08	1.93	0.678	1.37	4.86	0.0	1.08	3.07	2.02	7.2	0.21	0.0	
May-June	56	20	*	*	*	2.83	2.98	1.05	1.64	0.80	0.0	0.95	2.70	1.56	0.76	0.04	0.0	
July-Aug	63	8	*	*	*	1.42	1.67	0.586	1.10	1.48	0.0	0.85	2.42	1.88	2.5	0.18	0.0	
Sept-Oct	59	811	175.	57.9	87.8	21.9	5.44	0.633	1.25	1.55	7.99	4.03	34.6	1.97	2.4	0.0019	91.5	
Nov-Dec	59	830	133.	70.4	110.	42.7	-	1.03	2.06	3.53	3.11	-	41.4	2.00	3.4	0.0043	68.3	
Washington, D. C. - Lat. 38°59'N Long. 77°29'W Elev. 82 m																		
Jan-Feb	59	14	*	*	*	1.48	1.20	0.441	0.837	5.44	0.0	1.23	3.36	1.90	12.3	0.39	0.0	
Mar-Apr	60	21	*	*	*	2.40	2.47	0.738	1.56	3.47	0.0	0.97	3.25	2.11	4.7	0.17	0.0	
May-June	63	32	*	*	*	3.90	4.47	1.33	2.63	3.21	0.0	0.87	2.93	1.98	2.4	0.10	0.0	
July-Aug	58	16	*	*	*	1.56	1.39	0.530	1.11	2.18	0.0	1.12	2.94	2.09	4.1	0.14	0.0	
Sept-Oct	63	863	242.	97.1	117.	27.7	7.14	0.810	1.54	5.39	8.74	3.88	34.2	1.90	6.7	0.0062	120.	
Nov-Dec	56	1035	165.	131.	182.	66.0	-	1.49	2.83	1.81	2.50	-	44.3	1.90	1.2	0.0017	87.9	
Miami, Florida - Lat. 25°49'N Long. 80°17'W Elev. 4 m																		
Jan-Feb	61	19	*	*	*	2.03	1.73	0.550	0.833	1.91	0.0	1.17	3.69	1.52	3.5	0.17	0.0	
Mar-Apr	60	35	*	*	*	4.33	4.33	1.28	2.11	2.84	0.0	1.00	3.38	1.65	2.2	0.08	0.0	
May-June	62	22	*	*	*	3.13	3.14	0.964	1.82	2.27	0.0	1.00	3.25	1.89	2.4	0.10	0.0	
July-Aug	51	8	*	*	*	0.955	1.27	0.354	0.537	1.44	0.0	0.75	2.70	1.52	4.1	0.18	0.0	
Sept-Oct	63	499	169.	68.3	78.4	17.8	3.95	0.675	0.761	0.982	9.49	4.51	26.4	1.13	1.5	0.0020	101.	
Nov-Dec	56	1495	422.	182.	296.	102.	-	2.76	2.09	0.840	4.14	-	37.0	0.76	3.0	0.0006	65.9	
Mauna Loa, Hawaii - Lat. 19°28'N Long. 155°36'W Elev. 3394 m																		
Jan-Feb	59	11	*	*	*	1.22	1.11	0.384	0.676	1.51	0.0	1.10	3.18	1.76	3.9	0.14	0.0	
Mar-Apr	60	25	*	*	*	4.09	3.15	0.986	1.84	1.86	0.0	1.30	4.15	1.87	1.9	0.074	0.0	
May-June	63	27	*	*	*	3.18	2.83	1.10	2.00	2.44	0.0	1.12	2.89	1.82	2.2	0.090	0.0	
July-Aug	59	13	*	*	*	1.66	1.89	0.600	1.17	1.87	0.0	0.88	2.77	1.95	3.1	0.14	0.0	
Sept-Oct	53	133	25.3	11.1	12.0	3.50	1.26	0.323	0.609	1.12	7.23	2.78	10.8	1.89	3.5	0.0084	34.4	
Nov-Dec	56	1100	204.	140.	202.	66.9	-	1.69	2.88	0.577	3.05	-	39.6	1.70	0.34	0.0005	84.3	
Miraflores, Panama Canal Zone - Lat. 9°00'N Long. 79°35'W Elev. 10 m																		
Jan-Feb	59	36	2.00	1.03	1.04	1.23	0.954	0.302	0.529	0.753	1.63	1.28	4.07	1.75	2.5	0.021	3.4	
Mar-Apr	60	14	1.23†	*	*	1.11	1.26	0.346	0.554	1.33	1.1	0.88	3.21	1.60	3.8	0.095	0.0	
May-June	63	11	*	*	*	0.864	0.914	0.278	0.506	1.90	0.0	0.95	3.11	1.82	6.8	0.17	0.0	
July-Aug	59	3	*	*	*	0.186	0.223	0.068	0.109	0.763	0.0	0.83	2.74	1.60	11.2	0.25	0.0	
Sept-Oct	63	7	1.06†	0.49†	0.79†	0.165	0.070	0.041	0.093	0.423	6.4	2.36	4.02	2.27	10.3	0.06	12.0	
Nov-Dec	55	274	48.2	25.6	42.3	12.7	4.90	0.352	0.385	0.312	3.80	2.59	36.1	1.09	0.89	0.0011	72.7	
Guayaquil, Ecuador - Lat. 2°10'S Long. 79°52'W Elev. 7 m																		
Jan-Feb	53	5	(0.6)	(0.02)	*	0.283	0.488	0.085	0.191	2.25	(2.0)	0.58	3.33	2.29	26.	0.45	(0.2)	
Mar-Apr	59	5	*	*	*	0.235	0.228	0.081	0.202	2.69	0.0	1.03	2.90	2.49	33.	0.54	0.0	
May-June	33	4	*	*	*	0.250	0.230	0.086	0.159	1.80	0.0	1.09	2.91	1.85	21.	0.45	0.0	
July-Aug	56	2	*	*	*	0.099	0.126	0.047	0.095	1.55	0.0	0.79	2.11	2.02	33.	0.78	0.0	
Sept-Oct	63	3	(0.03)	*	*	0.199	0.268	0.101	0.208	1.54	(0.2)	0.74	1.97	2.06	15.2	0.51	0.0	
Nov-Dec	62	13	3.20	2.31	2.89	1.42	0.713	0.239	0.530	2.37	2.25	1.99	5.94	2.22	9.9	0.18	9.7	

TABLE 3 (Continued)
Radiochemical Analyses of Composite Bimonthly Air-Filter Collections During 1961

Month	Days** Sampled	Activity (dis/min per 100 std. cubic meters of air)†									Activity Ratios						
		Gross β	Ce ¹⁴¹	Sr ⁹⁰	Y ⁹¹	Ce ¹⁴⁴	Pm ¹⁴⁷	Sr ⁹⁰	Cs ¹³⁷	Pb ²¹⁰	Ce ¹⁴¹ / Ce ¹⁴⁴	Ce ¹⁴⁴ / Pm ¹⁴⁷	Ce ¹⁴⁴ / Sr ⁹⁰	Cs ¹³⁷ / Sr ⁹⁰	Pb ²¹⁰ / Sr ⁹⁰	Pb ²¹⁰ / Gross β	Sr ⁹⁰ / Sr ⁹⁰
Lima, Peru - Lat. 12°01'S Long. 77°07'W Elev. 30 m¶																	
Jan-Feb	51	4	*	*	*	0.420	0.336	0.156	0.286	0.551	0.0	1.25	2.69	1.83	3.5	0.14	0.0
Mar-Apr	60	3	*	*	*	0.240	0.198	0.097	0.193	0.177	0.0	1.21	2.47	1.99	1.8	0.06	0.0
May-June	63	6	*	*	*	0.518	0.449	0.208	0.206	0.825	0.0	1.15	2.49	0.99	4.0	0.14	0.0
July-Aug	52	6	*	*	*	0.568	0.552	0.248	0.378	0.705	0.0	1.03	2.29	1.52	2.8	0.12	0.0
Sept-Oct	61	7	*	*	*	1.09	1.20	0.520	0.850	0.633	0.0	0.96	2.21	1.63	1.2	0.09	0.0
Nov-Dec	64	6	0.67‡	0.59‡	0.84‡	0.669	0.535	0.248	0.442	0.470	1.00	1.25	2.70	1.78	1.9	0.08	2.4
Chacaltaya, Bolivia - Lat. 17°10'S Long. 68°15'W Elev. 5220 m																	
Jan-Feb	51	5	*	*	*	0.292	0.242	0.097	0.227	1.86	0.0	1.21	3.01	2.34	19.2	0.37	0.0
Mar-Apr	54	6	*	*	*	0.381	0.376	0.150	0.329	2.01	0.0	1.01	2.54	2.19	13.4	0.34	0.0
May-June	51	6	*	*	*	0.364	0.341	0.152	0.337	2.58	0.0	1.07	2.39	2.22	17.0	0.43	0.0
July-Aug	63	11	*	*	*	1.14	1.28	0.481	0.907	3.81	0.0	0.89	2.37	1.89	7.9	0.35	0.0
Sept-Oct	62	10	(0.42)	*	(0.21)	0.763	0.873	0.374	0.688	2.93	(0.6)	0.87	2.04	1.84	7.8	0.29	0.0
Nov-Dec	63	15	1.61‡	1.17‡	1.52‡	1.013	0.651	0.276	0.516	2.09	1.6	1.56	3.67	1.87	7.6	0.14	4.2
Antofagasta, Chile - Lat. 23°37'S Long. 70°16'W Elev. 519 m																	
Jan-Feb	43	12	*	*	*	1.41	1.17	0.507	0.765	1.35	0.0	1.21	2.78	1.51	2.7	0.11	0.0
Mar-Apr	60	8	*	*	*	0.950	0.910	0.349	0.744	3.45	0.0	1.04	2.72	2.13	9.9	0.43	0.0
May-June	60	8	*	*	*	0.798	0.791	0.310	0.612	3.87	0.0	1.01	2.57	1.97	12.5	0.48	0.0
July-Aug	52	7	*	*	*	1.02	1.10	0.413	0.740	0.989	0.0	0.93	2.47	1.79	2.4	0.14	0.0
Sept-Oct	63	8	(0.83)	*	*	1.31	1.41	0.564	1.14	0.503	(0.6)	0.97	2.32	2.02	0.89	0.06	0.0
Nov-Dec	63	16	1.26‡	0.90‡	1.31‡	1.57	1.57	0.590	1.067	0.595	0.8	1.00	2.66	1.81	1.01	0.037	1.5
Santiago, Chile - Lat. 33°27'S Long. 70°42'W Elev. 520 m																	
Jan§	25	13	*	*	*	1.97	1.78	0.708	1.44	1.28	0.0	1.11	2.78	2.03	1.8	0.10	0.0
Mar-Apr	-	-	No samples received			0.679	0.549	0.278	0.442	2.78	0.0	1.24	2.44	1.59	10.0	0.35	0.0
May§	31	8	Insufficient samples received														
July-Aug	-	-	Insufficient samples received														
Sept-Oct	63	7	*	*	(0.22)	0.972	1.41	0.418	0.908	0.813	0.0	0.69	2.33	2.17	1.9	0.12	0.0
Nov-Dec	63	10	(0.10)	(0.17)	(0.15)	0.908	1.22	0.494	1.004	1.25	(0.11)	0.74	1.84	2.03	2.5	0.13	(0.4)
Puerto Montt, Chile - Lat. 41°27'S Long. 72°57'W Elev. 5 m																	
Jan-Feb	-	-	No samples received														
Mar-Apr	-	-	Insufficient samples received														
May-June	-	-	Insufficient samples received														
July-Aug	54	4	*	*	*	0.465	0.481	0.215	0.311	0.390	0.0	0.98	2.16	1.45	2.7	0.15	0.0
Sept-Oct	63	4	*	*	*	0.504	0.614	0.261	0.294	0.273	0.0	0.82	1.93	1.13	1.05	0.07	0.0
Nov-Dec	63	4	*	(0.15)	*	0.640	0.784	0.349	0.532	0.175	0.0	0.82	1.83	1.52	0.50	0.04	(0.4)
Punta Arenas, Chile - Lat. 53°08'S Long. 70°53'W Elev. 3 m																	
Jan-Feb	47	4	*	*	*	0.489	0.429	0.185	0.370	0.161	0.0	1.14	2.64	2.00	0.87	0.04	0.0
Mar-Apr	60	3	*	*	*	0.405	0.424	0.177	0.346	0.036	0.0	0.96	2.29	1.95	0.20	0.012	0.0
May-June	44	3	*	*	*	0.268	0.236	0.108	0.210	0.157	0.0	1.14	2.48	1.94	1.45	0.05	0.0
Aug-Sept†	55	3	*	*	*	0.290	0.335	0.140	0.265	0.175	0.0	0.87	2.07	1.89	1.25	0.06	0.0
Oct†	35	2	*	*	*	0.217	0.333	0.134	0.263	0.146	0.0	0.65	1.62	1.96	1.09	0.07	0.0
Nov-Dec	56	2	(0.10)	(0.09)	*	0.186	0.249	0.111	0.224	0.126	(0.5)	0.75	1.68	2.02	1.14	0.06	(0.8)

*Activity not detectable at time of analysis

†All activities (except gross β) corrected for decay to midpoint of collection period. Counting error less than $\pm 2\%$ (standard deviation) unless otherwise indicated

‡Counting error estimated to be in range of 2-10% (standard deviation)

§Analysis of samples from a single month

¶Operation moved to this location from 12°06'S, 77°01'W, elev. 134 m on 3 July 1961

**Irregularities in numbers result from varying sampling schedule - initially 3 samples per week per station, later one 7-day collection per week

††Error made in grouping samples for analysis

() Value uncertain. Error exceeds $\pm 10\%$

season and decreases (minima) during the fall were observable in both hemispheres. Furthermore, the standard latitudinal profiles consisting of maxima in activity concentrations in the mid-latitudes of each hemisphere and a minimum near the equator were observable throughout the year. It is unfortunate that observations could not be made to determine if the normal spring maxima in the southern hemisphere had not approached the magnitude of those in the north, which would give an indication of approaching equilibrium in the stratospheric burdens over the two hemispheres.

The progressive changes that have occurred in the gross fission product concentrations in the air at Washington, D.C., Miami, Florida, and Antofagasta, Chile, during the period 1959-1961 are indicated in Fig. 2. The increases in activity during the spring season are evident at the two northern hemisphere sites but are less obvious at Antofagasta. The strong effect of the 1961 Soviet tests is evident in the hundredfold increase in radioactivity at Washington and Miami, with activity levels at those sites approximating that observed following the fall 1958 series of high yield nuclear tests in the Soviet Union.

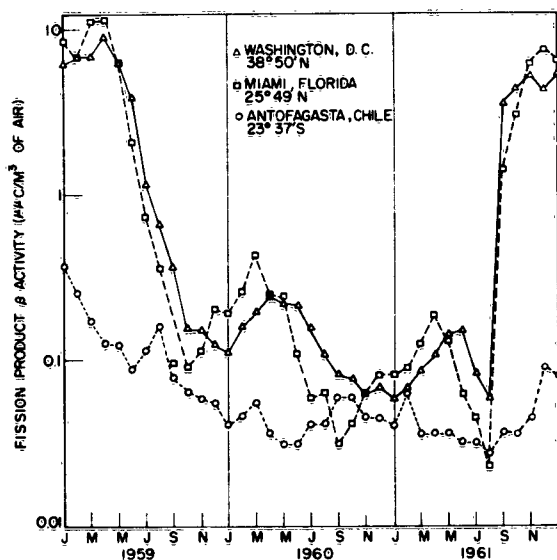


Fig. 2 - Progressive changes in the air concentration of fission products at Washington, Miami, and Antofagasta, 1959-1961

RADIOCHEMICAL ANALYSES

The results of the radiochemical analyses of composite bimonthly samples from each of the

stations are reported in Table 3 in units of disintegrations/minute per 100 standard cubic meters of air. Also recorded are some of the activity ratios which are of current interest. All radioactivity results (except for the gross β measurements) have been corrected for decay to the midpoint of the collection period.

Sr^{90} IN THE AIR AT GROUND LEVEL

The Sr^{90} concentrations in the air at various sites in the northern hemisphere during 1961 (prior to the 1961 Soviet tests) were generally lower than those found in 1960 but the relative rate of decrease was significantly lower than during the preceding year, 1959-1960. The large stratospheric source over the arctic region generated by the Soviet test series of 1958 had been dissipated at a rapid rate during the spring of 1959 with the result that deposition in succeeding years was primarily from a stratospheric reservoir over the tropical region. There must, therefore, exist either a rather large tropical reservoir from which fallout occurs at a much lower rate (i.e., several years vs. less than one year for arctic debris) or intrusion of radioactivity into the lower stratosphere from debris injected extremely high into the atmosphere (i.e., Teak and Orange shots of the U.S. Hardtack series) must be occurring at such a rate that the total available stratospheric burden of Sr^{90} has remained relatively unchanged. Other investigators have reported the presence of this high altitude debris at higher levels in the northern and southern polar stratospheres (9, 10); some has also been detected at ground level (11).

The changes in the Sr^{90} concentrations that have occurred at four sites in the North Temperate Zone during the period January 1959 through February 1962 are shown in Fig. 3. The spring maxima in stratospheric deposition are evident in each of the years 1959, 1960, and 1961, with relatively little difference indicated between the latter two years. The great increase in the Sr^{90} concentration following the onset of renewed nuclear testing by the Soviet Union in September 1961 is apparent, with Sr^{90} levels approaching those of the 1958-1959 period. In spite of the high concentrations of gross activity appearing at these sites during the fall and winter of 1961, the bulk of the Sr^{90} and other long-lived isotopes did not appear until the usual time for the 1962 spring peak in fallout. This would indicate, as expected from the reported size of the weapons tested, that the bulk of the fission products was directly introduced into the stratosphere and that the radioactivity appearing at early times along the 80th meridian was from debris which were primarily tropospheric in origin.

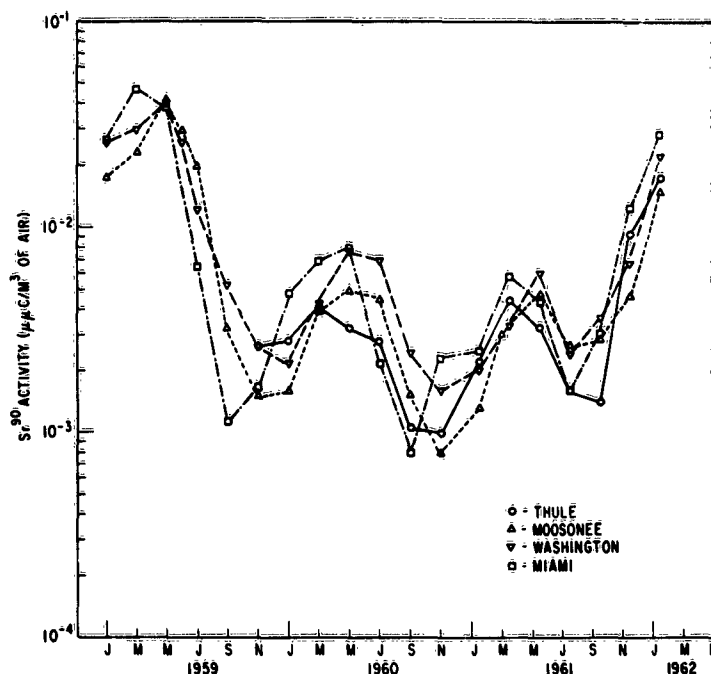


Fig. 3 - Concentrations of Sr^{90} in the air of the North Temperate Zone, 1959-1961

In the southern hemisphere, due to the small quantity of transequatorial transport of debris from the 1961 Soviet nuclear test program, it was possible to continue to observe throughout 1961 and early 1962 the changes in Sr^{90} concentrations from older sources of contamination. Here, in contrast to trends observed north of the equator, there has been a gradual increase in Sr^{90} concentrations indicating an increase in the stratospheric burden of radioactivity over the southern hemisphere or else its migration to a position where downward mixing into the troposphere progresses more rapidly. Probably both mechanisms are contributing to this increase in the ground level concentration of Sr^{90} .

As indicated in Fig. 4, there are clearly defined spring maxima in Sr^{90} levels at Antofagasta during both 1960 and 1961, while the maxima at Punta Arenas are less obvious and are displaced in time toward the summer season. (This effect of latitude on the time of appearance of the maximum is well-established in the northern hemisphere (4,7).) The generally ill-defined maxima in the more southerly latitudes is perhaps directly related to the absence of any appreciable reservoir of fission debris in the antarctic stratosphere, though a contributing factor could be a less effective exchange across the antarctic tropopause than across the arctic tropopause. The abrupt increase in the Sr^{90} concentra-

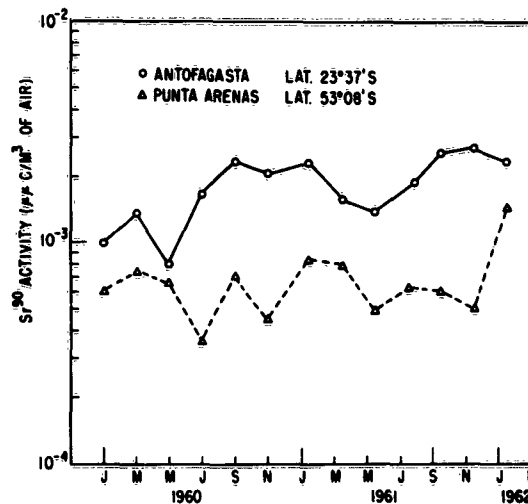


Fig. 4 - Concentrations of Sr^{90} in the air at Antofagasta and Punta Arenas, Chile, 1960-1961

tion at Punta Arenas during early 1962 (also noted at Puerto Montt and Santiago) could be the result of the appearance of Teak and Orange debris in the ground level air following its migration into the antarctic region at high altitudes and its subsequent subsidence during the southern hemisphere

spring; some objections to this assignment are discussed later. The presence of debris from these tests has been noted by Gustafson (12), who detected in some of the 1959-1960 Santiago samples the Rh^{102} tracer characteristic of the Orange high altitude test at Johnston Island in August 1958.

A comparison of the concentrations of Sr^{90} in the air at corresponding high altitude sites north and south of the equator (Mauna Loa and Chacaltaya) during 1960-1961 is shown in Fig. 5. At both of these sites the expected seasonal variations in activity concentration are observed, roughly six months out of phase, together with a trend of decreasing activity in the northern hemisphere and increasing activity in the southern hemisphere. The increase in Sr^{90} from the fall 1961 USSR tests is evident at Mauna Loa in the November-December 1961 and January-February 1962 collections.

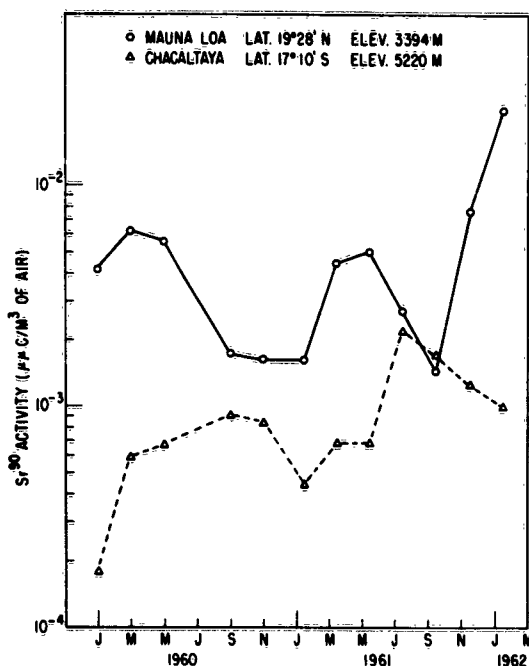


Fig. 5 - Concentrations of Sr^{90} in the air at corresponding high altitude sites north and south of the equator

Pb^{210} IN THE AIR AT GROUND LEVEL

Pb^{210} (RaD) is a constituent of the normal atmosphere and results from the successive decay of radon and its daughters in the air after diffusion of radon gas from the soil. The total quantity of this isotope in the atmosphere must remain rather invariant with time, but its concentration in the

air at any one site would be expected to be governed by the geographical location of the site and the meteorology of the area.

The Pb^{210} has been measured in air filter collections from the 80th meridian stations during the past several years, but insufficient data has been obtained to document seasonal changes in this isotope, though large month to month variations have been observed. There is a definite latitude dependence to the Pb^{210} concentration, as indicated in Fig. 6, which resembles that observed for Sr^{90} .

The unsymmetrical shape results from the fact that more radon, and hence more Pb^{210} , is produced in the northern hemisphere due to its larger land area, while the tropical minimum is perhaps related to rainfall patterns there. The lower concentrations observed at the higher latitudes are again related to the lack of radon sources in these areas, due to a minimum of exposed land surfaces and the depletion of Pb^{210} by the various deposition mechanisms before its arrival at these sites.

During 1961 the Pb^{210} content of the air was generally much higher than its Sr^{90} content at all sites, as indicated in Table 3 and by the Sr^{90} and Pb^{210} profiles shown in Fig. 6. The resumption of large scale nuclear testing will undoubtedly cause

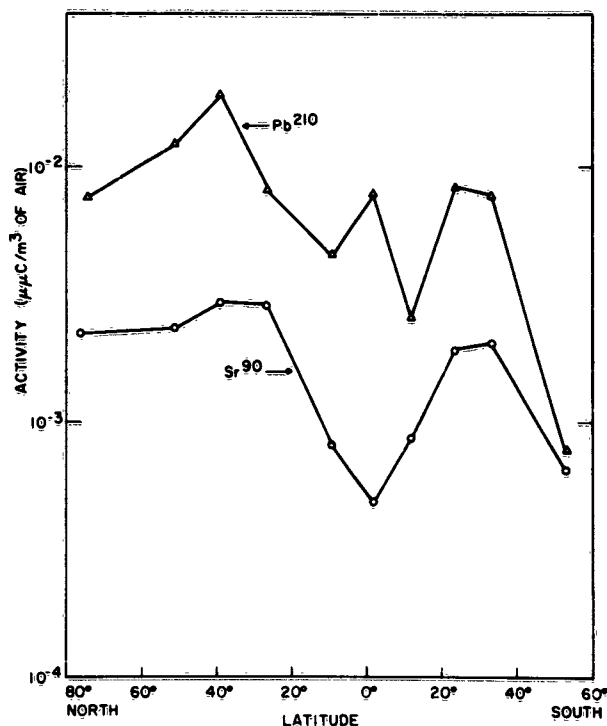


Fig. 6 - Profiles of the average Pb^{210} and Sr^{90} concentrations in the air during the period September 1960-August 1961

an increase in the Sr^{90} levels during the coming year to near their 1958-1959 values at which time the Sr^{90} concentration will exceed that of Pb^{210} . The increased Sr^{90} concentration at sites in the northern hemisphere resulting from the 1961 USSR tests has already been noted in collections made during the period January-April 1962.

ACTIVITY RATIOS

The variations in the relative concentrations of certain radioactive isotopes with time and with location can give an indication of the apparent age of fission debris appearing at the different sites, or what is generally more important, these ratios can indicate whether the sources of debris at different collection sites are the same or different. Age determinations have more significance when they involve the shorter-lived isotopes present in the debris from recent nuclear explosions; at later times, when only such longer-lived isotopes as Ce^{144} , Pm^{147} , and Sr^{90} are present, dating of the source of the debris cannot be done with any accuracy but differences between collections can be related to different sources of debris. This latter con-

dition existed throughout 1961 until the onset of nuclear testing in the USSR supplied a source of short-lived isotopes as active tracers of debris from this test series.

$\text{Ce}^{144}/\text{Sr}^{90}$

Plots of the ratios of Ce^{144} to Sr^{90} activity in the air at several sites of comparable distance north and south of the equator during the period 1958-1961 are shown in Fig. 7. The fast depletion of debris from the 1958 Soviet fall series of nuclear tests in the northern hemisphere air is evident in the more rapid decrease in the $\text{Ce}^{144}/\text{Sr}^{90}$ ratio than could be accounted for by radioactive decay alone. No comparable change occurred in the southern hemisphere where the contribution of USSR debris was negligible. By early 1960 the age difference between the radioactivity in the two hemispheres, as determined from this ratio, had decreased to two to three months, with the younger debris lying in the north. This age difference was maintained throughout 1960 and 1961 up until the time the fresh USSR debris appeared on the scene.

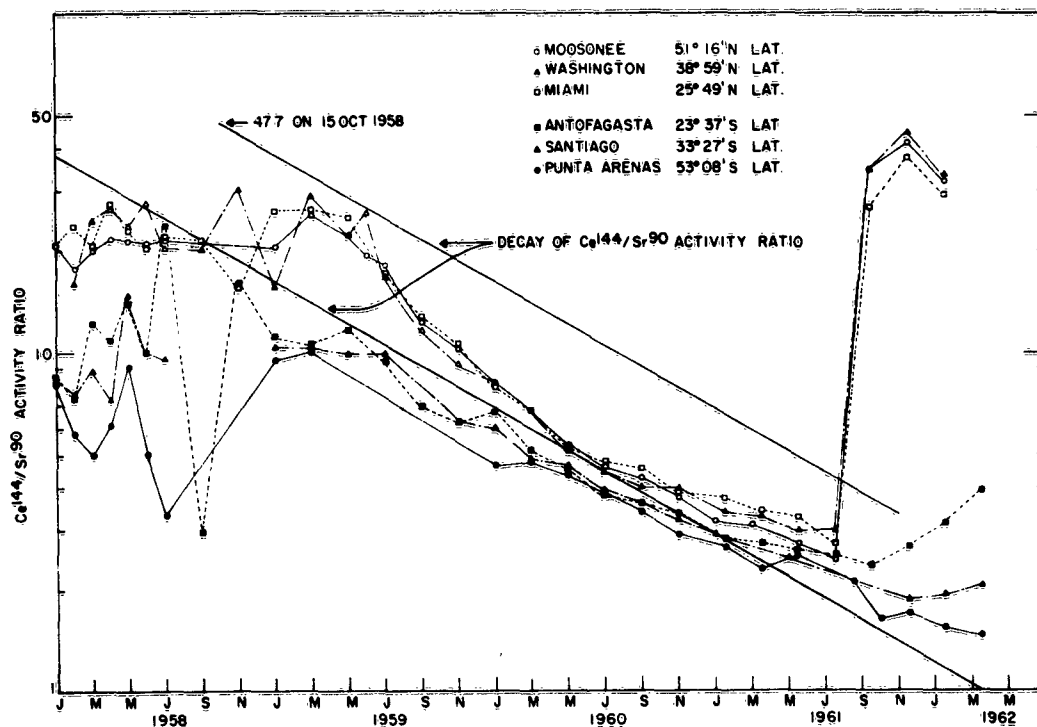


Fig. 7 - Ratios of Ce^{144} to Sr^{90} activity at various sites during the period 1958-1961

TABLE 4
Calculated Contribution of Possible Sources of Younger Debris to Total Sr^{90}
in Ground-Level Air During November 1960 and July-August 1961*

	Sr^{90} from younger source (percent of total)		
	French test Feb. 13, 1960	Teak-Orange tests August 6, 1958†	Fall 1958 Soviet tests Oct. 15, 1958†
November 1960			
Northern Hemisphere	2.3	14.6	10.8
Southern Hemisphere	0.9	5.2	4.0
July-August 1961			
Northern Hemisphere	7.6	49.5	36.0
Southern Hemisphere	6.2	36.2	27.3

*Based on observed changes in $\text{Ce}^{144}/\text{Sr}^{90}$ ratio; ratio assumed to be 47.7 at zero time for all shots (15).

†Effective midpoint of series.

Beginning in mid-1960 there was a gradual change in the slope of the $\text{Ce}^{144}/\text{Sr}^{90}$ decay curve which was suggestive of the simultaneous appearance in both hemispheres of nuclear debris of more recent origin than that then being collected. Since the older debris dated from an apparent detonation time in late 1957 (which could represent a composite of debris from Hardtack, Grapple, and prior test series), the source of the younger debris could have been from either the Hardtack or Grapple tests, or less likely, the USSR tests of 1958. The magnitude of the change is such that it could not be accounted for by the relatively low yield French nuclear tests.

Though the presence of debris from the high-altitude shots of the Hardtack series (Teak and Orange) over Johnston Island has been documented in both polar stratospheres and at ground level, there is no definite evidence that this is the source of the fresher debris. Since the debris from these tests has been noted to exist primarily in the polar regions of the stratosphere, the time of appearance at ground level should be related to the pronounced seasonal changes in downward mixing from these regions and thus be approximately six months out of phase in the two hemispheres. There might also be expected to occur, after the observed decrease in rate of decay, a subsequent increase as polar deposition became less important and relatively more debris from the stable tropical source appeared. This, too, does not appear to be happening. Finally, the large increase in Sr^{90} concentration in the air at Punta Arenas during Janu-

ary-February 1962, which might have been indicative of stratospheric deposition of Teak-Orange debris, was not accompanied by the corresponding increase in the $\text{Ce}^{144}/\text{Sr}^{90}$ ratio at that site which would have been expected from such an influx of younger debris.

While the source of debris having the higher $\text{Ce}^{144}/\text{Sr}^{90}$ debris cannot be identified with any degree of certainty, it is definite that the source is not a nuclear test of more recent origin than 1958; no short-lived fission products other than those produced in the French Sahara tests, have been observed and these could have contributed little to the stratospheric inventory of Ce^{144} . For sufficient Ce^{144} to have been introduced into the stratosphere to cause the observed changes in the $\text{Ce}^{144}/\text{Sr}^{90}$ ratio would have required 2 to 3% of the Sr^{90} appearing at ground level in the Northern Hemisphere in November 1960 and about 9% of that appearing in July-August 1961 to have come from the French test of February 13, 1960, having an estimated yield of only 60-70 kilograms. Kuroda (13) has suggested that some of the French debris did enter the stratosphere but estimated that the stratospheric reservoir of Sr^{90} was enhanced by at most a few tenths of one percent.

The estimated contribution of some of the possible sources of younger debris to the total Sr^{90} content of the air at ground level during November 1960 and July-August 1961, based on the observed enhancement of the $\text{Ce}^{144}/\text{Sr}^{90}$ ratio, is presented in Table 4. As indicated earlier, the French test could not have contributed the quantity of Sr^{90} required

by this analysis; similarly, it would be extremely unlikely that debris from the 1958 Soviet test series would be contributing substantially equally to activity levels in the two hemispheres by August 1961. This would require a faster rate of mixing than has been observed and is not compatible with the observed differences in absolute air concentrations of Sr^{90} in the two hemispheres. In spite of other discrepancies noted above, the seemingly only possible source of the younger debris can be the U.S. Hardtack series of nuclear tests, presumably the Teak and Orange shots. If this is true, this source must have contributed about 10% of the Sr^{90} observed in the ground level air during November 1960 and nearly 50% of that observed in July-August 1961, the last period at which a determination could be made. These results are in agreement with those of Walton and Fried (14) based on an analysis of the $\text{Ce}^{144}/\text{Sr}^{90}$ ratios found in rainwater collections at Westwood, New Jersey, and with those of the High Altitude Sampling Program (HASP) (9).

$\text{Cs}^{137}/\text{Sr}^{90}$

The variability of the $\text{Cs}^{137}/\text{Sr}^{90}$ activity ratio during 1961 was quite similar to that observed during 1960. The median value of the 73 measurements was 1.88 with 50% of the values falling in the range 1.89 ± 0.14 . This may be compared with the corresponding range of 1.90 ± 0.15 for the 73 measurements made in 1960. The mean value during 1961 was 1.81; its displacement from the median was due to a secondary grouping of ratios near 1.52 together with a few scattered extremely low values. In both 1960 and 1961 the general patterns were the same with Miami, Miraflores, and Puerto Montt exhibiting $\text{Cs}^{137}/\text{Sr}^{90}$ ratios consistently below the 50% grouping around the median.

The possibility of loss of Cs^{137} during the ashing process prior to addition of Cs carrier cannot be overlooked. However, it is difficult to understand why test runs did not exhibit any loss of Cs or why such losses would consistently occur in samples from a few sites, when all samples had been treated in as nearly identical manner as possible. Since no unusual ratios of other isotopes relative to Sr^{90} were found, it is evident that fractionation of Cs^{137} relative to Sr^{90} has occurred either in nature or in the sampling and analytical process.

Other Activity Ratios

The $\text{Ce}^{144}/\text{Pm}^{147}$ ratio continued to decrease as the shorter-lived Ce^{144} became depleted relatively faster than Pm^{147} due to radioactive decay. The

influx of fresh debris in the latter part of 1961, however, added more Ce^{144} activity than Pm^{147} to the atmosphere causing a marked increase in this ratio. Since this ratio is not very sensitive to changes in the composition or age of the fission product mixture, other activity ratios have been employed to describe the movement of fresh debris from the 1961 Soviet tests. This is discussed in the following section.

SPREAD OF DEBRIS FROM THE USSR TESTS OF SEPTEMBER-NOVEMBER 1961

At the time of resumption of nuclear testing by the Soviet Union on September 1, 1961, there existed no background of short-lived fission products in the atmosphere. The last fresh activity had been found only at Miraflores and Guayaquil in early 1961 as a result of the December 27, 1960 French test in the Sahara desert; no debris from the April 1961 French test was detected at any of the 80th meridian sites. The short lived isotopes Ce^{141} , Sr^{89} , and Y^{91} observed in the latter part of 1961 were thus due solely to the USSR tests and served to document the intrusion of debris from this source into areas where fresh debris had been absent for a long period of time.

Gross β measurements indicated the arrival of fresh debris in quantity at all northern hemisphere 80th meridian sites except Miraflores during the week of September 12-18, 1961; Miraflores did not intercept any identifiable quantity of activity until mid-November. The gross β measurements were not sufficiently specific to indicate the transequatorial passage of debris during 1961, except to Guayaquil in the latter part of December. Observed increases in activity could not be assigned unambiguously to these tests since this period is the expected time for the seasonal increase in stratospheric fallout in the southern hemisphere. Radiochemical analyses on the other hand did show definite penetration of USSR debris as far south as Antofagasta during November-December with indications that traces of it appeared as far south as Punta Arenas.

An indication of the relative quantities of fresh debris is given in Fig. 8 where the profiles of Ce^{141} (33.1 day half-life), Sr^{89} (50.5 day half-life), and Sr^{90} (27.7 year half-life) concentrations in the air at various latitudes along the 80th meridian are shown. The highest concentrations of Ce^{141} and Sr^{89} in the Southern Hemisphere are at Guayaquil and in the neighborhood of Chacaltaya and Antofagasta, where they are less than 1% of those observed at similar latitudes in the north. Trans-equatorial mixing to this extent is not unexpected

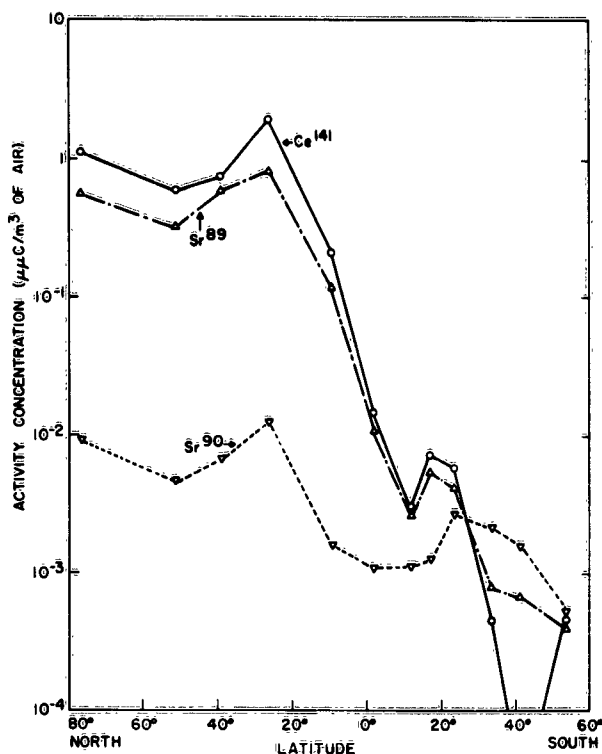


Fig. 8 - Profiles of the activity concentrations of Ce^{141} , Sr^{89} and Sr^{90} in the air at ground level along the 80th meridian during November-December 1961

since a similar transfer of debris from the first French test in the Sahara desert at 27° north latitude to the southern hemisphere was observed (7). The mechanism by which northern hemisphere debris appear in higher concentrations in the region 17-23°S than at 12°S (Lima) is suggested whereby debris penetrating to the vicinity of the equator are entrained in the rising air masses which subsequently move away from the equator and descend to ground level in the subtropical region, thus effectively by-passing the Lima area. The quantity of Ce^{144} , Pm^{147} , Sr^{90} , and Cs^{137} associated with the traces of fresh activities in the southern hemisphere were not sufficient to alter appreciably the trends observed in the ratios involving these long-lived isotopes.

FUTURE STUDIES OF ATMOSPHERIC RADIOACTIVITY

A number of scientific studies relating to the size of the stratospheric reservoir of radioactive material, the rate of transfer across the equator and the rate of movement into the troposphere

have been disrupted by the introduction of fresh debris into the stratosphere. The new sources of activity currently being produced in both the arctic and tropic regions are not point sources in either time or location, and, being composed of similar distributions of fission products, afford no means for identification of their origin. Furthermore, the background of older debris is so completely masked that it is no longer of value as a tracer for stratospheric or tropospheric air movements.

During 1962 and for several years after any future agreement on cessation of nuclear testing, the information obtained from air monitoring programs might be expected to parallel that obtained during the period of active testing in 1958 and the moratorium on nuclear testing in effect until September 1961.

While these sources of activity are available efforts should be continued to document the concentrations of airborne radioactivity encountered in the various areas together with the fallout in these areas, and particularly to make studies of the mechanisms by which deposition takes place. With the levels of activity to be expected in the atmosphere during the next year or so, the study of the size distribution of radioactive particles and its variation with time and weather become relatively more simple to perform than in recent years.

CONCLUSIONS

The fission product content of the tropospheric air in the northern hemisphere continued, through August 1961, the downward trend observed since the peak activity was recorded in the spring of 1959, with strong seasonal increases and decreases associated with a spring maximum in downward mixing of radioactive debris from a stratospheric reservoir. In the southern hemisphere seasonal variations were not so pronounced and a trend toward increasing air concentrations of long-lived fission products was observed, suggestive of southward mixing of debris across the equator into the southern stratosphere. The influx of fresher debris, presumed to be from the high altitude shots of the Hardtack series, into both hemispheres was indicated by changes in the Ce^{144}/Sr^{90} activity ratio.

The resumption of nuclear testing by the Soviet Union in September 1961, while forcing a termination of those studies dealing with the background of old radioactive fission products in the atmosphere, did provide a large source of shorter-lived isotopes as radioactive tracers for meteorological processes. Through use of this fresh tracer, by the end of 1961 the maximum concentration of USSR debris in the southern hemisphere was observed to be less than one percent of that in the northern hemisphere, again confirming the presence of a strong equatorial barrier to the southward mixing of tropospheric debris.

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phere, and a tendency toward equilibration of the activity con-
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north of the equator and an increase south of the equator. Prior
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After September 1961, the fission product radioactivity in the
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